

## N O T I C E

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Final Report of

Study of Surface Phenomena at Low Temperatures

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## I. Introduction

The study of superfluid helium in finite geometries is important for both fundamental and technological reasons. In the former category is the basic question of the relation between superfluidity and Bose-Einstein condensation. Although the latter is ruled out<sup>1</sup> for a finite thickness ( $D$ ) film at finite temperature  $T$ , superfluidity is nevertheless observed in numerous experiments.<sup>2</sup> This presents a major dilemma for our understanding of this phenomenon of superfluidity in any dimensionality. In any case, it leaves (at best) only the Landau two-fluid model as a basis for predicting behavior of helium confined in a finite geometry. This aspect has represented the focus of our work; our results<sup>3,4</sup> have suggested that space experiments can test this idea.

From the technical point of view, size-constrained helium can be utilized as an ideal system for testing concepts of nucleation; this has such practical applications as weather prediction. Our research has recently led to suggest useful experimental tests in this domain.

This project summary will direct attention in turn to three problems concerning which successful results have emerged during the year's investigation: the liquid-vapor interface, films and droplets, and ion-induced clusterings. A considerable part of the work remains incomplete. Some will be reported in future publications. Work is continuing in the second and third area (not funded, however).

## II. Results

### a. Liquid -Vapor Interface

The results of this investigation have already been published,<sup>3</sup> so only a brief summary follows.

If one studies capillary waves (oscillations of the liquid-vapor interface), one finds that an infinite planar boundary is stable only in the presence of a gravitational field  $g$ .<sup>5,6</sup> A long wavelength divergence as  $g \rightarrow 0$  can be prevented alternatively by letting the lateral size  $L_0$  be finite. Since the divergence is logarithmic in  $g$  and  $L_0$ , macroscopic terrestrial experiments are of little value for probing this phenomenon. Our work<sup>3</sup> has determined the behavior expected for a general fluid (i.e., from extreme quantum to extreme classical) as a function of the variables  $g$  and  $L_0$ . The numerical results indicate that space experiments can fruitfully test these notions. As described in the paper,<sup>3</sup> fundamental questions of the interfacial diffuseness<sup>7</sup> and how to interpret computer simulation results<sup>8</sup> can be usefully explored. We have not, however, considered in detail the problems of experimental design.

### b. Films and Droplets

The conventional approach to treating physisorbed films generates the so-called Frenkel-Halsey-Hill relation between  $D$  and the vapor saturation ratio  $P/P_0$ . This is only an approximation, however, because of the implicit assumption that the film does not differ substantially from bulk fluid.<sup>9</sup> For a thin film, this errs because of both non-uniformity introduced by the substrate and explicit size-dependent effects. We have neglected the former for simplicity. By the latter is meant the inclusion of interfacial waves (e.g., ripples) and a proper accounting of the normal modes

of the film. For example, in a thick film<sup>10</sup> the elementary excitations have wave vector  $k_z = n\pi/2D$ . The result of evaluating the thermodynamic properties will thus depend on D. For example, a film on a surface of area A has a phonon contribution to the free energy given by<sup>11</sup>

$$\beta F_{ph} = (A/2\pi) \int_0^{\infty} dk K \sum_{k_z} \ln (1 - e^{-\lambda k}) ,$$

where K is the wave vector (k) component parallel to the substrate and  $\lambda = hs/k_B T$  is the thermal phonon wavelength (s is the speed of sound). The sum in this expression reduces to an integral when D → ∞. For finite thickness, there is a correction when D ≤ λ = 18 Å K for <sup>4</sup>He. This is an explicit size effect which is amenable to experimental verification; specifically the chemical potential  $\mu \propto (\partial F/\partial D)$  is equal for the film and vapor in equilibrium. Our calculations indicate the effect be a few percent for T = 1 K when λ = D (larger for smaller D or T). Because of the attractive effect of the substrate (density enhancement), it is unrealistic to extend this calculation to very small D with confidence.

The ideal system for studying size effects is the droplet, in which case neither gravitational nor substrate-induced van der Waals potentials play a role (g is unimportant if the radius R is small compared to the capillary length  $(\sigma/\rho g)^{1/2} \sim 1$  mm for helium). We have analyzed the low T thermodynamic properties in terms of the Landau hydrodynamic model. What emerges is a situation similar to that found for planar film geometry; size effects appear significantly only when R ≤ λ<sub>ph</sub> ~ 20 Å. Because the answer is so disappointing (i.e., inaccessible except at very low T) we do not belabor the details, which will appear in a future publication (co-authored by Dr. S. Rauber).

### c. Ion-Induced Clustering

Ions have been known for at least a century<sup>12</sup> to be nucleation centers for condensation. Their advantage over surfaces in this regard is merely one of ideal simplicity of characterization; there are no uncertainties analogous to surface imperfection, for example.

We have begun a small effort in this area designed to understand clustering of the atoms about an ion immersed in a low T fluid. This has been stimulated partly by a resurgence of activity,<sup>13-16</sup> but also by a realization that ions in He are a particularly fruitful probe of nucleation.<sup>17</sup>

The basic point of our work is that the range of possible impurity ions represent a variable source of the clustering process.<sup>18</sup> The static and dynamic properties of the core region can then be altered in a controllable way and the consequences measured experimentally. This prospect is very exciting; we are collaborating with Akinci and Northby at the University of Rhode Island in this study.

### III. Summary and Future Prospects

We have barely scratched the surface of research problems in this area. Much remains to be done in the domain of analysis of possible experiments. Given the limited (one year) duration of the project, the incomplete development is not surprising. We believe that the study of interfacial diffuseness suggested in Ref. 3 is a potentially useful low (or zero) gravity experiment. Most of our other work provides background for future attention.

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